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Evolution of quantum criticality in the system $CeNi_9Ge_4$

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Abstract. The heavy fermion system $CeNi_9Ge_4$ exhibits a paramagnetic ground state with remarkable features such as: a record value of the electronic specific heat coefficient in systems with a paramagnetic ground state, $\gamma = C/T \simeq 5.5$ J/mol K^2 at 80 mK, a temperature-dependent Sommerfeld–Wilson ratio, $R = \chi/\gamma$, below 1 K and an approximate single ion scaling of the $4f$-magnetic specific heat and susceptibility. These features are related to a rather small Kondo energy scale of a few Kelvin in combination with a quasi-quartet crystal field ground state. Tuning the system towards long range magnetic order is accomplished by replacing a few at.% of Ni by Cu or Co. Specific heat, susceptibility and resistivity studies reveal $T_N \sim 0.2$ K for $CeNi_8CuGe_4$ and $T_N \sim 1$ K for $CeNi_8CoGe_4$. To gain insight whether the transition from the paramagnetic NFL state to the magnetically ordered ground state is connected with a heavy fermion quantum critical point we performed specific heat and ac susceptibility studies and utilized the μSR technique and quasi-elastic neutron scattering. [1]

1. Introduction

Some exciting novel electronic phenomena in solids, e.g. high-temperature and heavy-Fermion superconductivity, have been observed when tuning strongly correlated electron systems by substitution doping, pressure or other non-thermal parameters from a symmetry breaking ground state towards a symmetry conserving one, e.g. from long range magnetic order towards a paramagnetic Fermi liquid ground state. Prominent examples in this context are pressure induced, magnetically mediated unconventional superconducting states in antiferromagnetic (AF) Kondo lattice systems $CePd_2Si_2$, $CeIn_3$ [1] and in ferromagnetic UGe_2 [2]. If continuous, such symmetry breaking phase transition at virtually zero temperature gives rise to quantum critical behavior at finite temperature (for review see e.g. [3]) which e.g. manifests in the non-Fermi liquid (NFL) behavior of various heavy Fermion systems [4, 5].

$CeNi_9Ge_4$ is a very interesting paramagnetic Kondo lattice system that exhibits remarkable features such as a record value of the electronic specific heat coefficient with a paramagnetic ground state, $\gamma = C/T \simeq 5.5$ J/mol K^2 at 80 mK [6], which is the highest among the strongly

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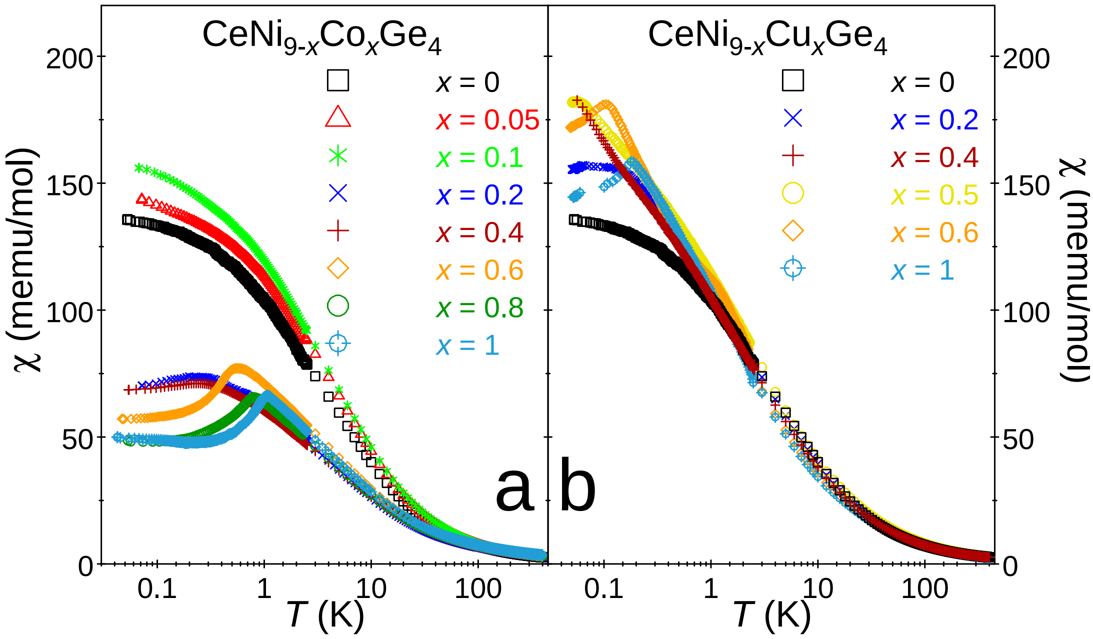

Figure 1. The magnetic susceptibility $\chi(T)$ of $CeNi_{9-x}Co_xGe_4$ (a) and $CeNi_{9-x}Cu_xGe_4$ (b) in semi-logarithmic plots (redrawn from [13, 14]). AFM transitions are evident for $x > 0.5$.

correlated electron systems. $CeNi_9Ge_4$ further exhibits a strongly temperature-dependent Sommerfeld–Wilson ratio, $R = \chi/\gamma$, below 1 K and an approximate scaling of the $4f$-magnetic specific heat and susceptibility for the Ce concentration in a magnetically dilute solid solution $Ce_xLa_{1-x}Ni_9Ge_4$ [7]. Such scaling of the magnetic contributions to the specific heat and susceptibility reveals that the single ion type interactions, such as crystal field (CF) and Kondo effects, are responsible for the physical properties of these compounds. The CF ground state of $CeNi_9Ge_4$ has been determined by means of single crystal susceptibility measurements which were analyzed in terms of the tetragonal crystal field model in combination with a Kondo screening correction by the poor mans scaling approach yielding a CF level scheme with a ground state formed by two doublets ($\Gamma_7^{(1)}$, $\Gamma_7^{(2)}$) split by about 0.5 meV and a well separated Γ_6 doublet with an excitation energy of 11 meV [8]. The low-temperature Kondo energy scale referring to the quasi-quartet ground state which results from the analysis of the macroscopic static susceptibility as well as microscopic neutron studies of the quasi-elastic linewidth is about 0.3 meV [8]. The splitting of the $\Gamma_7^{(1)}$ and $\Gamma_7^{(2)}$ doublets is, thus, of comparable magnitude as the Kondo energy scale and thereby smeared to a quasi-quartet ground state which shows up in the specific heat as roughly $C \sim -T \ln T$, which is a typical behaviour observed in NFL systems at low temperature [6, 7].

Based on the above estimates of CF and Kondo energy scales of $CeNi_9Ge_4$ numerical renormalization group calculations using the $SU(4)$ Anderson impurity model accounted for the basic thermodynamic features of $CeNi_9Ge_4$ reasonably well and demonstrated that the temperature dependent Sommerfeld–Wilson ratio $R(T) = \chi(T)/\gamma(T)$ results from an $SU(2)$ to $SU(4)$ cross-over [9, 10].

Cerium Kondo lattice systems having Kondo temperatures T_K as low as a few Kelvin usually exhibit a magnetically ordered ground state [11], i.e. they are dominated by RKKY inter-site interactions. $CeNi_9Ge_4$ with $T_K \sim 3.5$ K and no magnetic frustration (tetragonal structure with easy direction of magnetisation in c-axis), however, displays a paramagnetic ground state. A factor in favor of a paramagnetic ground state is the effectively quasi-fourfold ground state degeneracy as proposed in the theoretical model of Coleman [12]. In this model above a critical value of the Kondo coupling constant, $(J\rho)_c$, the spin-compensated Kondo-lattice ground state is stable and this value $(J\rho)_c$ is shown to tend to zero as $O(1/N)$, providing new justification of applicability of the Kondo lattice model for rare earth based strongly correlated electron systems [12]. To explore the significance of RKKY interactions in $CeNi_9Ge_4$ and to check for the possibility of quantum criticality in this system, we have studied Ni-site substitutions by Cu and Co, i.e. equivalent to electron and hole doping, respectively. The aim to tune the system towards long range magnetic order is in fact accomplished by replacing a few at.% of Ni by Co as well as by Cu (see figure 1 and references [13, 14] for further details including specific heat and resistivity studies). It is surprising to find magnetic order for both, $CeNi_8CoGe_4$ and $CeNi_8CuGe_4$, because electron and hole doping are expected to drive the Kondo temperature in opposite directions. The latter is in fact supported by the susceptibility data displayed in figure 1 where Co substitution in 1a clearly reduces the susceptibility [14], i.e. indicating an increase of T_K, whereas Cu substitution in 1b tends to increase the low temperature susceptibility, thus, pointing towards a reduction of T_K in the solid solution $CeNi_{9-x}Cu_xGe_4$ [13]. What both solid solutions, $CeNi_{9-x}T_xGe_4$ with $T =$ Co and Cu, show in common is a reduction of the effectively quasi-fourfold degeneracy of the CF ground state of the parent $CeNi_9Ge_4$ compound towards a two-fold degenerate one in both $CeNi_8CoGe_4$ and $CeNi_8CuGe_4$ which is proposed by the analysis of magnetic entropy data [13, 14].

In this paper we present new microscopic studies of magnetic correlations in $CeNi_{9-x}Cu_xGe_4$ by means of quasi-elastic neutron scattering and muon spin relaxation (μSR) experiments. Macroscopic studies of thermodynamic properties such as magnetic susceptibility, specific heat and thermal expansion revealed a quantum critical point in this solid solution series where $CeNi_{8.6}Cu_{0.4}Ge_4$ exhibits quantum critical behavior with χ and $C/T \propto -\ln T$ as well as a strongly composition dependent behavior of the Grüneisen ratio $\mathit{\Gamma}(T) \propto \alpha(T)/C(T)$ at low temperatures where $\mathit{\Gamma}(T \to 0)$ increases by orders of magnitudes right at the critical composition $CeNi_{8.6}Cu_{0.4}Ge_4$ [13].

2. Experimental details

Polycrystalline samples used for neutron scattering and μSR studies namely $CeNi_9Ge_4$, $CeNi_{8.6}Cu_{0.4}Ge_4$, $CeNi_8CuGe_4$, $Ce_{0.8}La_{0.2}Ni_9Ge_4$, and $LaNi_9Ge_4$ were synthesized by high frequency induction melting on a water cooled copper hearth under a protective argon atmosphere. The starting materials were Ce and La metals (Ames MPC [15], 99.95%), Ni (Johnson-Matthey, GB, 99.999%) and zone-refined Ge ingots (Johnson-Matthey, GB, 99.9999%). In a first step, Ni and Ge were melted together to produce a master alloy, which was then alloyed with Ce metal. The samples were finally annealed for one week at 1270 K in evacuated quartz ampules. Standard x-ray diffraction performed on powder revealed essentially phase pure samples crystallizing in the tetragonal space group $I4/mcm$ where substitution of Ni by Cu leads to a linear volume increase reaching 0.8% for $CeNi_8CuGe_4$ as compared to $CeNi_9Ge_4$. Details of the structural characterization were reported earlier [13].

Cold neutron quasi-elastic scattering experiments were carried out on the IN6 time-of-flight spectrometer at ILL Grenoble. The spectrometer was operated with an incident neutron energy of 3.15 meV (wavelength $\lambda = 5.12$ Å), yielding an energy resolution of 70 μeV at full width half maximum (FWHM). Powder samples, each with masses near 20 g, were enclosed in flat Al-can sample holders yielding an effective sample thickness of about 1.3 mm which was taken into account for proper absorption corrections. For calibration we measured a vanadium reference sample of the same disc-shape geometry at 2 K. For subtracting the background signal of the sample holder and intrinsic non-magnetic contributions to the total scattering (mainly phononic ones), we measured $LaNi_9Ge_4$ reference sample at same experimental conditions and

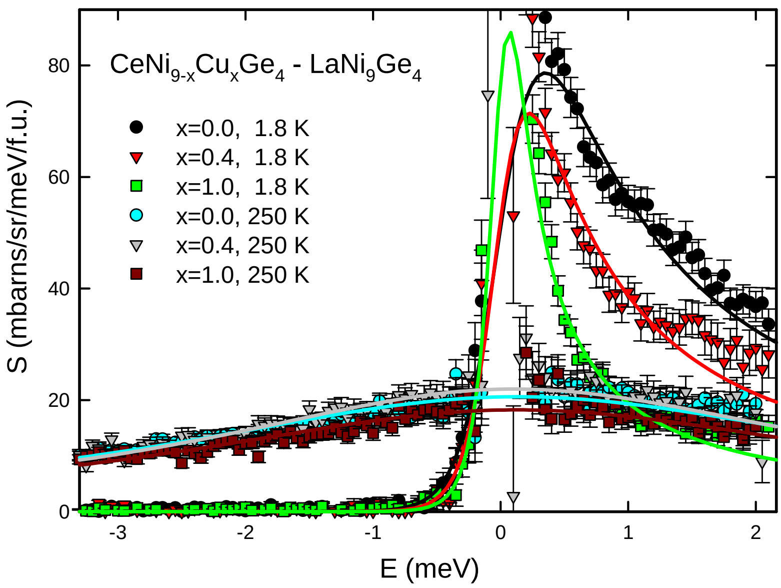

Figure 2. The magnetic correlation function $S_m(\omega)$ at 1.8 K and 250 K for compositions $CeNi_{9-x}Cu_xGe_4$ as labeled; solid lines are Lorentzian fits according to equation 2.

temperatures.

The muon spin relaxation (μSR) experiments at temperatures down to 35 mK were performed on the *MuSR* spectrometer at the ISIS facility where pulses of muons are implanted into the sample at 50 Hz and with a FWHM of 70 ns. These muons are thermalized in the bulk of the sample within few ps and decay with a half-life, $\tau_\mu = 2.2\,\mu$s into positrons, which are emitted preferentially in the direction of the muon spin axis. Each positron is time stamped and therefore the muon spin polarisation which corresponds to the asymmetry between positron counts in the forward and backward detectors (in so-called longitudinal geometry) can be followed as a function of time. In the present experiments, an initial asymmetry of 0.28 refers to an initially perfect spin polarization of the implanted muons parallel to the beam forward direction.

3. Cold neutron quasi-elastic studies

The variation of the Kondo temperature in the solid solution $CeNi_{9-x}Cu_xGe_4$ is evaluated from the quasi-elastic paramagnetic response probed by cold neutron scattering, i.e. via measurements of correlation functions $S(q,\omega)$ as described earlier in reference [8]. As the coherent and incoherent cross sections of $CeNi_{9-x}Cu_xGe_4$ with $x \leq 1$ and $LaNi_9Ge_4$ match each other within a few percent, the magnetic scattering $S_m(q,\omega)$ of cerium is obtained by subtracting the $LaNi_9Ge_4$ data representing phonon plus the background contributions. The magnetic scattering data $S_m(q,\omega,T \geq 1.8\text{K})$ obtained thereby exhibit only a weak q-dependence of the intensity which corresponds well to the usual Ce^{3+} form factor. Accordingly, q-integrated (section from 0.4 to 1.0 Å^{-1}) magnetic correlation functions $S_m(\omega)$ of $CeNi_9Ge_4$, $CeNi_{8.6}Cu_{0.4}Ge_4$, and $CeNi_8CuGe_4$ measured at 1.8 K and 250 K are displayed in figure 2. The magnetic scattering,

$$S_m(\omega, T) = \frac{\chi''(\omega, T)}{1 - \exp(-\hbar\omega/k_B T)}, \tag{1}$$

is related to the absorptive component $\chi''(\omega, T)$ of the dynamic susceptibility which in case of a normal paramagnetic response with a single exponential decay of the magnetisation density is given by a Lorentzian

$$\chi''(\omega,T) = \chi'(T)\frac{\omega\Gamma(T)}{\omega^2+\Gamma^2(T)} \tag{2}$$

where $\Gamma(T)$ and $\chi'(T)$ are the temperature dependent relaxation rate (line width) and static susceptibility, respectively. Despite of an underlying CF splitting, the quasi-elastic response is acceptably well described by simple Lorentzian fits according to equation 2. While all the high temperature data measured at 250 K where $k_{\mathrm{B}}T$ is larger than the overall CF splitting are essentially on top of each other, there are rather significant changes of the low temperature quasi-elastic response.

The Lorentzian fits indicated by solid lines in figure 2 reveal a reduction of the line width Γ(1.8K) from 0.47 meV to 0.36 meV and 0.20 meV for $CeNi_9Ge_4$, $CeNi_{8.6}Cu_{0.4}Ge_4$, and $CeNi_8CuGe_4$, respectively. Simultaneously, also the overall integrated intensity which corresponds to the static susceptibility χ'(1.8K) (see equation 2) decreases from 0.104 emu/mol to 0.088 emu/mol, and 0.073 emu/mol, respectively. Although the trend is similar the significant variation of χ'(1.8K) seems in contradiction with the SQUID susceptibility data in figure 1b showing a minor variation of χ(1.8K). A close agreement of the value proposed by the Lorentzian fit with the SQUID result is observed for $CeNi_9Ge_4$ (from CF parameters in reference [8] 96% of the total magnetic response is expected from the $\Gamma_7^{(1)}$-$\Gamma_7^{(2)}$ quasi-quartet), but an increasing lack of quasi-elastic intensity as compared to the static SQUID susceptibility in the case of $CeNi_{8.6}Cu_{0.4}Ge_4$, and $CeNi_8CuGe_4$. The latter is obviously connected with a growing splitting of the $\Gamma_7^{(1)}$ and $\Gamma_7^{(2)}$ doublets caused by Cu substitution which alters the local environment of the cerium ions and, thereby, changes their CF scheme. The quasi-elastic neutron data, thus, reveal that Ni/Cu substitution causes a significant reduction of the Kondo energy scale and also a reduction of the effective ground state degeneracy towards a doublet in the case of $CeNi_8CuGe_4$. The latter is also confirmed by inelastic neutron data measured recently with an incident neutron energy of $E_{\mathrm{i}} = 20.5$ meV at FRMII where an approximate CF scheme with doublets at ground state, 5 meV and 12 meV is observed for $CeNi_8CuGe_4$ [16].

4. Muon spin relaxation studies

In order to probe magnetic correlations at low (dilution fridge) temperatures we utilized the μSR technique which is a local probe method sensitive to extremely small internal fields and ideally suited to detect any kind of static magnetism as well as dynamic magnetic features (if accessible in μs time scale of the muon life time). The μSR technique has, thus, been extensively applied to heavy fermion systems (see e.g. the review by Amato [17]).

The zero-field μSR measurements down to about 40 mK for $CeNi_9Ge_4$, $CeNi_{8.6}Cu_{0.4}Ge_4$, $CeNi_8CuGe_4$, and a magnetically dilute sample $Ce_{0.8}La_{0.2}Ni_9Ge_4$ are displayed in figure 3 as time dependent depolarization of the muon spins, i.e. asymmetry estimated from the positions detected in beam forward and beam backward directions. All relaxation data measured at highest temperatures displayed in figure 3, i.e. at 4 − 5 K, are in approximate agreement with the Gaussian Kubo-Toyabe relaxation function (see e.g. reference [17]) which refers a depolarisation of the muon spins by quasi-static nuclear dipolar fields. As the largest nuclear moments are contributed by copper atoms, $CeNi_8CuGe_4$ in figure 3a clearly displays the quickest relaxation at high temperatures and nuclear dipolar fields maintain their dominant role down to 0.3 K. Just near 200 mK, where the susceptibility in figure 1b exhibits a sharp cusp indicating the onset of AF order, there is a dramatic change of the μSR signal showing a rather quick depolarization towards a typical background asymmetry $A_{\mathrm{bg}} \sim 0.05$ at low temperatures. The absence of any sign of coherent frequency oscillations in the zero field μSR spectra which has been observed also in some other antiferromagnetic materials, e.g. $Ce_8Pd_{24}Ga$ [19], refers to a muon stopping site in a high symmetry position with respect to the Ce sublattice where dipolar fields of AF

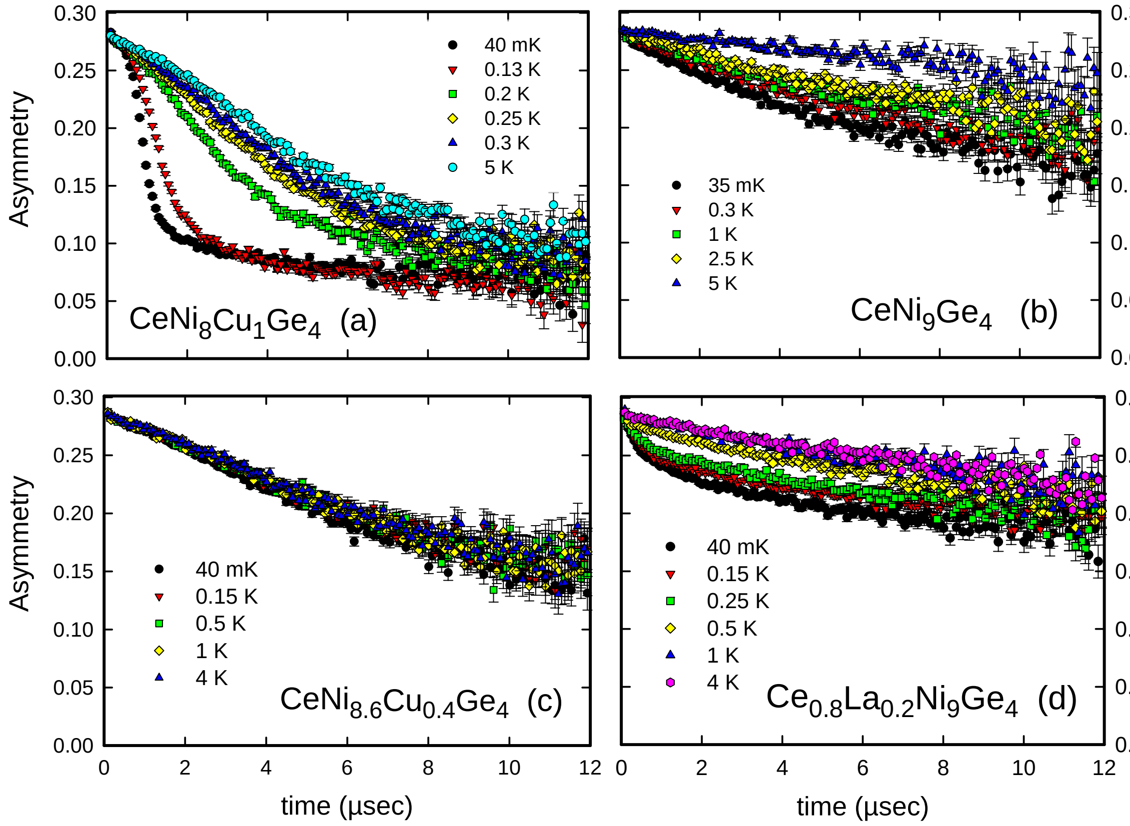

Figure 3. The time dependent μSR asymmetry data of $CeNi_8CuGe_4$ (a), $CeNi_9Ge_4$ (b), $CeNi_{8.6}Cu_{0.4}Ge_4$ (c), and $Ce_{0.8}La_{0.2}Ni_9Ge_4$ (d) at selected temperatures and zero field.

ordered Ce moments may compensate each other to zero. In the presence of substitutional disorder in $CeNi_8CuGe_4$, muon spins at such point of compensating dipolar fields should, of course, experience some distribution of static internal fields with a zero mean at the muon site. This interpretation of the $CeNi_8CuGe_4$ 40 mK data is supported by a markedly different field dependence of $CeNi_8CuGe_4$ observed in longitudinal field μSR measurements (not shown for the sake of brevity) as compared to all other compounds studied in this investigation.

The exponential low temperature relaxation of $CeNi_9Ge_4$ and magnetically dilute $Ce_{0.8}La_{0.2}Ni_9Ge_4$ in figure 3b and 3d refers a muon spin depolarization by dynamically fluctuating dipolar fields. In the case of $CeNi_9Ge_4$ a clean simple exponential behaviour $A(t) \propto \exp(-\lambda t)$ with a depolarization rate $\lambda = 0.133\,\mu\mathrm{s}^{-1}$ at 35 mK is observed (see figure 3b). As already discussed in reference [8] a simple exponential μSR signal rules out a disorder or inhomogeneity dominated NFL regime which would be indicated by a broad distribution and a strong temperature dependence of μSR relaxation rates [18]. The latter situation seems to apply for the solid solution $Ce_{0.8}La_{0.2}Ni_9Ge_4$ which exhibits a significantly larger initial relaxation rate and strongly stretched exponential behaviour. The larger initial relaxation rate is counter-intuitive for the magnetically dilute case in particular when keeping in mind the approximate concentration scaling of magnetic contributions to the specific heat and susceptibility reported by Killer *et al.* [7]. These effects of magnetic dilution are even more remarkable when considering the corresponding results of $CeNi_{8.6}Cu_{0.4}Ge_4$ which according to our thermodynamic studies [13] is located right at the critical concentration for the onset of AF order and clearly exhibits quantum critical behavior with χ and $C/T \propto -\ln T$. Against all expectations μSR results in figure 3c do not reveal any temperature dependence of the $CeNi_{8.6}Cu_{0.4}Ge_4$ data even from 40 mK to 4 K where muon spins essentially probe nothing else than the nuclear dipolar fields of the copper cores.

The only plausible interpretation of these puzzling results are strong AF short range magnetic correlations causing a compensation dipolar fields at the muon site which is most effective right at the quantum critical point, i.e. muon will not see any change with temperature because nuclear dipolar fields remain dominant at all temperatures. Magnetic dilution through Ce/La substitution of course creates at least two kinds of muon stopping sites: (i) in between a pair of Ce-ions where correlated $4f$ moments may compensate each other and (ii) in between a pair of Ce and La where the muon spin feels the full Ce moment. Longitudinal field μSR data (not shown) in fact support this scenario where a smaller number of muon sites ($\leq 40\%$) is of the second type and causes the quick initial relaxation and is more robust against the externally applied longitudinal field, i.e., connected with larger local fields at the muon site, and a larger number of muon sites is of type (i) displaying a much slower relaxation which is easily suppressed by just a few 10 G longitudinal field similar to the field dependence seen in $CeNi_9Ge_4$ and $CeNi_{8.6}Cu_{0.4}Ge_4$.

5. Summary

The effectively quasi-fourfold ground state degeneracy in combination with a Kondo temperature of only a few Kelvin places the paramagnetic heavy fermion system $CeNi_9Ge_4$ in an exceptional position among Kondo lattice systems where the paramagnetic ground state could be stabilized by the quasi-quartet CF ground state. The latter is suggested by the fact that tuning $CeNi_9Ge_4$ towards a magnetic ground state is accomplished by Ni/Cu as well as Ni/Co substitution (equivalent to electron and hole doping, respectively) which are both leading to a slightly modified local environment of the cerium ions and, thereby, cause a reduction of the CF ground state towards a doublet one. These modifications of ground state degrees of freedom are revealed by microscopic studies via cold neutron quasi-elastic scattering on $CeNi_9Ge_4$ and the solid solutions $CeNi_{8.6}Cu_{0.4}Ge_4$ and $CeNi_8CuGe_4$ where a significant narrowing of the quasi-elastic line at low temperatures and simultaneously some loss in quasi-elastic intensity is observed. The latter indicates a transfer of quasi-elastic to inelastic scattering caused by a splitting of the quasi-quartet CF ground state towards two well separated doublets.

Local probe μSR studies performed down to temperatures near 40 mK revealed rather unexpected results where on the one hand, magnetic dilution (Ce/La substitution) tends to increase the local fields experienced by the implanted muons, whereas on the other hand, tuning towards quantum criticality in $CeNi_{8.6}Cu_{0.4}Ge_4$ tends to annihilate the dipolar fields of cerium moments at the muon stopping sites. A cancellation of the dipolar fields of cerium $4f$ moments is only possible if muons locate in a high symmetry position with respect to the Ce sublattice where strong AF correlations lead to a pairwise compensation of cerium dipolar fields. The μSR results are, thus, suggestive of strong AF short range dynamic correlations being present in the unusual Kondo lattice ground state of $CeNi_9Ge_4$.

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